JC20 Rec'd PCT/PTO SEP 2 0 2001

FORM PTO-1390 (REV 10-95) U.S DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE		ATTORNEY'S DOCKET NUMBER MERCK 2306
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. §371		U S. APPLICATION NO. (If known, see 37 CFR §1.5) 09/937011
INTERNATIONAL APPLICATION NO. PCT/EP00/02009	INTERNATIONAL FILING DATE 8 MARCH 2000	PRIORITY DATE CLAIMED 24 MARCH 1999

TITLE OF INVENTION

APPARATUS AND PROCESS FOR THE PREPARATION OF SALT MELTS, AND THE USE THEREOF

APPLICANT(S) FOR DO/EO/US

OHREM, Hans, Leonhard, et al.

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. [x] This is a **FIRST** submission of items concerning a filing under 35 U.S.C. §371.
2. [ ] This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. §371.
3. [ ] This express request to begin national examination procedures (35 U.S.C. §371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. §371(b) and PCT Articles 22 and 39(1).
4. [x] A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. [x] A copy of the International Application as filed (35 U.S.C. §371(c)(2))
   a. [ ] is transmitted herewith (required only if not transmitted by the International Bureau).
   b. [x] has been transmitted by the International Bureau.
   c. [ ] is not required, as the application was filed in the United States Receiving Office (RO/US).
6. [x] A translation of the International Application into English (35 U.S.C. §371(c)(2)).
7. [x] Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. §371(c)(3))
   a. [ ] are transmitted herewith (required only if not transmitted by the International Bureau).
   b. [ ] have been transmitted by the International Bureau.
   c. [ ] have not been made; however, the time limit for making such amendments has NOT expired.
   d. [x] have not been made and will not be made.
8. [ ] A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. §371(c)(3)).
9. [x] An oath or declaration of the inventor(s) (35 U.S.C. §371(c)(4)).
10. [ ] A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. §371(c)(5)).

Items 11. to 16. below concern document(s) or information included:

11. [ ] An Information Disclosure Statement under 37 C.F.R. §§1.97 and 1.98.
12. [ ] An assignment document for recording. A separate cover sheet in compliance with 37 C.F.R. §§3.28 and 3.31 is included.
13. [x] A FIRST preliminary amendment.
    [ ] A SECOND or SUBSEQUENT preliminary amendment.
14. [ ] A substitute specification.
15. [ ] A change of power of attorney and/or address letter.
16. [ ] Other items or information:

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U.S. APPLICATION NO. (if known, see 37 CFR §1.5) 09/937011	INTERNATIONAL APPLICATION NO PCT/EP00/02009	ATTORNEY'S DOCKET NUMBER MERCK 2306

17. ☒ The following fees are submitted:

BASIC NATIONAL FEE (37 CFR §1.492 (a) (1) - (5)):

		CALCULATIONS	PTO USE ONLY
Search Report has been prepared by the EPO or JPO	$860.00		
International preliminary examination fee paid to USPTO (37 CFR §1.482)	$690.00		
No international preliminary examination fee paid to USPTO (37 CFR §1.482) but international search fee paid to USPTO (37 CFR §1.445(a)(2))	$710.00		
Neither international preliminary examination fee (37 CFR §1.482) nor international search fee (37 CFR §1.445(a)(2)) paid to USPTO	$1000.00		
International preliminary examination fee paid to USPTO (37 CFR §1.482) and all claims satisfied provisions of PCT Article 33(2)-(4)	$100.00		
ENTER APPROPRIATE BASIC FEE AMOUNT =		$860.00	
Surcharge of **$130.00** for furnishing the oath or declaration later than ☐ 20 ☐ 30 months from the earliest claimed priority date (37 C.F.R. §1.492(e)).			

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		
Total claims	11 - 20 =	0	x $ 18.00	$0.00	
Independent claims	1 - 3 =	0	x $ 80.00	$0.00	
MULTIPLE DEPENDENT CLAIM(S) (if applicable)			+ $ 270.00		

TOTAL OF ABOVE CALCULATIONS =	$860.00	
Reduction of 1/2 for filing by small entity, if applicable. A Verified Small Entity Statement must also be filed (Note 37 C.F.R. §§1.9, 1.27, 1.28).		
SUBTOTAL =	$860.00	
Processing fee of **$130.00** for furnishing the English translation later than ☐ 20 ☐ 30 months from the earliest claimed priority date (37 C.F.R. §1.492(f)).		
TOTAL NATIONAL FEE =	$860.00	
Fee for recording the enclosed assignment (37 C.F.R. §1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 C.F.R. §§3.28, 3.31). $40.00 per property.		
TOTAL FEES ENCLOSED =	$860.00	
	Amount to be refunded:	
	charged:	

a. ■ A check in the amount of $860.00 to cover the above fees is enclosed.

b. ☐ Please charge my Deposit Account No. 13-3402 in the amount of $ ______ to cover the above fees. A duplicate copy of this sheet is enclosed.

c. ■ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 13-3402. A duplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 C.F.R. §§1.494 or 1.495 has not been met, a petition to revive (37 C.F.R. §1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO: Customer Number 23,599

23599
PATENT TRADEMARK OFFICE

SIGNATURE

Anthony J. Zelano
NAME

27,969
REGISTRATION NUMBER

Filed: 20 SEPTEMBER 2001

AJZ:kmo

IN THE UNITED STATES DESIGNATED/ELECTED OFFICE

International Application No.	:	PCT/EP00/02009
International Filing Date	:	8 MARCH 2000
Priority Date(s) Claimed	:	24 MARCH 1999
Applicant(s) (DO/EO/US)	:	OHREM, Hans Leonard, et al.

Title: APPARATUS AND PROCESS FOR THE PREPARATION OF SALT METALS, AND THE USE THEREOF

PRELIMINARY AMENDMENT

Commissioner for Patents
Washington, D.C. 20231

SIR:

Prior to calculating the national fee, and prior to examination in the National Phase of the above-identified International application, please amend as follows:

IN THE CLAIMS:

3. (Amended) Apparatus according to Claim 1, characterized in that solids metering units (2,3) for controlled addition of mixing of the starting materials are arranged upstream of the stirred reactor (1).

4. (Amended) Apparatus according to Claim 1, characterized in that a purification unit (5,6), consisting of a column or tower (5) filled with metal granules (d) and a column or tower filled with alkali metal salt (MX), is arranged downstream of the tubular reactor (4).

5. (Amended) Process for the preparation of salt melts of the general formula in which

M is Li, Na, K, Rb or Cs,

D is Al, Ga, In or Tl, and

X is F, Cl, Br or I,

by reacting a metal halide of the formula DX_3 (II) with an alkali metal salt of the formula MX (III), characterized in that the reaction is carried out in an apparatus according to Claim 1, where the reaction is carried out firstly in a stirred reactor (1) and subsequently in a tubular reactor (4).

7. (Amended) Process according to Claim 5, characterized in that the salts are reacted at temperatures between 50 and 800°C.

8. (Amended) Process according to Claim 5, characterized in that the reaction is carried out continuously.

9. (Amended) Use of salts of the formula (I), and prepared by a process according to Claim 5, as melt electrolyte in electrochemical cells, batteries, sodium batteries and primary batteries.

10. (Amended) Use of salts of the formula (I), and prepared by a process according to Claim 5, as storage medium in heat stores and as heat-transfer medium.

11. (Amended) Use of salts of the formula (I), and prepared by a process according to Claim 5, for blanketing and purifying molten metals and for electrocoating materials.

REMARKS

The purpose of this Preliminary Amendment is to eliminate multiple dependent claims in order to avoid the additional fee. Applicants reserve the right to reintroduce claims to canceled combined subject matter.

Attached hereto is a marked-up version of the changes made to the claims by the current amendment. The attached pages are captioned "**Version With Markings to Show Changes Made**".

Respectfully submitted,

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AJZ:jmm

FILED: 20 SEPTEMBER 2001

VERSION WITH MARKINGS TO SHOW CHANGES MADE

Claims 3-5 and 7-11 have been amended as follows:

3. (Amended) Apparatus according to Claim 1~~ or 2~~, characterized in that solids metering units (2,3) for controlled addition of mixing of the starting materials are arranged upstream of the stirred reactor (1).

4. (Amended) Apparatus according to ~~one of Claims~~Claim 1~~ to 4~~, characterized in that a purification unit (5,6), consisting of a column or tower (5) filled with metal granules (d) and a column or tower filled with alkali metal salt (MX), is arranged downstream of the tubular reactor (4).

5. (Amended) Process for the preparation of salt melts of the general formula in which

M is Li, Na, K, Rb or Cs,

D is Al, Ga, In or Tl, and

X is F, Cl, Br or I,

by reacting a metal halide of the formula DX_3 (II) with an alkali metal salt of the formula MX (III), characterized in that the reaction is carried out in an apparatus according to ~~Claims~~Claim 1~~ to 5~~, where the reaction is carried out firstly in a stirred reactor (1) and subsequently in a tubular reactor (4).

7. (Amended) Process according to Claim 5~~ or 6~~, characterized in that the salts are reacted at temperatures between 50 and 800°C.

8. (Amended) Process according to ~~one of Claims~~Claim 5~~ to 7~~, characterized in that the reaction is carried out continuously.

9. (Amended) Use of salts of the formula (~~I~~I), ~~defined in Claim 1~~ and prepared by a process according to ~~Claims~~Claim 5~~ to 8~~, as melt electrolyte in electrochemical cells, batteries, sodium batteries and primary batteries.

10. (Amended) Use of salts of the formula (~~1~~I), ~~defined in Claim 1~~ and prepared by a process according to ~~Claims~~Claim 5~~ to 8~~, as storage medium in heat stores and as heat-transfer medium.

11. (Amended) Use of salts of the formula (~~1~~I), ~~defined in Claim 1~~ and prepared by a process according to ~~Claims~~Claim 5~~ to 8~~, for blanketing and purifying molten metals and for electrocoating materials.

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Merck Patent Gesellschaft
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Apparatus and process for the preparation of salt melts, and the use thereof

Apparatus and process for the preparation of salt melts, and the use thereof

The invention relates to an apparatus and a process for the preparation of salt melts and mixtures thereof by means of a tubular reactor and a continuous stirred reactor, in which the starting materials are melted and brought to reaction, and the reaction products are subsequently passed through columns for purification.

Melts of salts, such as, for example, $NaAlCl_4$, have various areas of application. Salt melts can be employed as storage medium in heat stores, as heat-transfer agents, for example in heating baths, for blanketing and purifying molten metals, for electro-coating of high-melting materials or as melt electrolyte in primary batteries, as described in GB 2,046,506. A further possible application of these salts is in rechargeable sodium batteries. The salts are employed in batteries which have operating temperatures of between 130°C and 200°C (K.M. Abraham, D.M. Pasquariello, J. Electrochem. Soc., Vol. 137, 1189-1190 (1990)).

DE 3419279 describes an electrochemical cell in which the cathode matrix is impregnated with a sodium/aluminium halide salt melt electrolyte.

A relatively new area of application is the "ZEBRA battery". This high-temperature cell consists of an electrode of liquid sodium, a beta-aluminium electrolyte and an electrode of transition-metal chloride in an $NaAlCl_4$ melt (B. Cleaver, V.S. Sharivker, J. Electrochem. Soc., Vol. 142, 3409-3413 (1995)).

DE 3718920 describes the preparation of salt melts by adding a pure metal and an alkali metal halide to the melt. The reaction cell is operated above the melting

point of the salt melt. In the working example, the alkali metal halide is NaCl, the molten alkali metal is sodium, and the separator is beta-aluminium oxide. Owing to the use of pure sodium, special safety precautions, such as working under a protective-gas atmosphere, must be taken. The reactions must take place in separate cells, since poisoning of the separator by the by-product $AlHal_3$ formed must be prevented.

All the processes disclosed hitherto for the preparation of salt melts operate batchwise. A batch procedure has some severe disadvantages compared with a continuous preparation process. During a batch change, the apparatus must be opened. The product can then be contaminated by the oxygen from the ambient air, water and dust. The batch change results in down times of the plant and thus in a reduced space-time yield. An effective discontinuous process requires large apparatuses. The start-up process requires correspondingly more energy and time. It is been found that, in particular during start-up of the plants, impurities can be introduced into the process. FR 2168912 describes a complex purification process for alkali metal halogen aluminates. The 2-step purification process consists of oxygen treatment for degrading the organic impurities and aluminium treatment for precipitating iron and heavy metals. The aluminium treatment must be carried out under a nitrogen or argon atmosphere.

For the preparation of the alkali metal halogen aluminates, the reaction of corresponding aluminium halides and alkali metal halides in a sealed tube is described (Friedman, Taube, J. Am. Chem. Soc., 72, 2236-2243 (1950)). In this process, an increase in pressure to 6 - 7 atmospheres is observed, which results in problems (FR 2168912). The apparatuses must be fitted with the appropriate safety precautions.

For continuous processing of the salts, basically tubular reactors and continuous stirred reactors are suitable.

Tubular reactors are ideal reactors for the continuous processing of liquid media. The residence-time distribution here is very narrow, which means that all particles have approximately the same residence time, and a very narrow quality distribution is thus produced. A high surface area/volume ratio and turbulent flow mean that a tubular reactor is highly suitable for heat transfer. For use for a solid, however, the prerequisite of flowability is lacking. In addition, heat transfer for melting a salt mixture is very poor owing to the poor thermal conduction of the granular starting materials, which results in very long residence times.

Another possibility for carrying out reactions continuously is the continuous stirred reactor. Here, the granulated starting materials can be metered into the stirred melt. This facilitates convective heat transfer, which considerably accelerates heating of the starting materials. In particular if subliming salts are to be used as starting materials, a temperature below the sublimation temperature is necessary in order to avoid an excess pressure in the system. A severe disadvantage of the continuous stirred reactor is the residence-time distribution. This results in a significant proportion of unreacted starting materials always being present in the product discharge stream.

The object of the invention is to provide a continuous process for the preparation of pure salt melts which excludes the disadvantageous effects of the environment, minimizes the energy requirement and facilitates an optimum space-time yield. A further

object is to make large amounts of salt melts available in the shortest possible time.

The object according to the invention is achieved by an apparatus for the preparation of salt melts, and mixtures thereof, of the general formula

$$MDX_4 \quad (I)$$

in which

M is Li, Na, K, Rb or Cs,

D is Al, Ga, In or Tl, and

X is F, Cl, Br or I,

essentially consisting of a heatable stirred reactor (1) and a downstream tubular reactor (4), where the stirred reactor (1) contains a zone which, owing to the tank geometry, cannot contain solids, and the tubular reactor (4) or its feed line extends into this solid-free zone.

The invention furthermore relates to a process for the preparation of salt melts, and mixtures thereof, of the general formula (I) by reacting a metal halide of the formula DX_3 (II) with an alkali metal salt of the formula MX (III) in the abovementioned apparatus.

The process products are suitable for use as melt electrolyte in electrochemical cells, as storage medium in heat stores, as heat-transfer agent, for example in heating baths, for blanketing and purifying molten metals, for electrocoating of high-melting materials or as melt electrolyte in rechargeable sodium batteries and primary batteries.

Surprisingly, it has been found that a suitable combination of a continuous stirred reactor and a plurality of tubular reactors allows a salt melt of the desired quality to be produced continuously.

The process can be carried out using all continuous and heatable stirred reactors which appear suitable to the person skilled in the art. For the task of stirring, use can be made of all suitable stirrers, such as propeller, inclined blade, disc, impeller, cross-blade, gate or blade stirrers. The stirrer should facilitate good heat transfer between the reactor wall and the salt or melt.

Stirred reactors are usually made of steel. The aggressive salt melts can attack this material in a corrosive manner.

It has been found that stirred reactors whose main components are made of nickel alloys are particularly suitable for the processing of salts and salt melts. The relatively low mechanical stress on the reactor shell means that nickel can also be used as the tank material. It is also possible to employ reactors made of glass. It has furthermore been found that the metal parts of the stirred reactor which come into contact with the salts or salt melts can be protected against corrosive and abrasive damage by surface coatings with materials known to the person skilled in the art, such as PTFE/PFA, enamel or ceramic materials.

In order to improve the efficiency of the stirred reactor, it is necessary that no solid enters the reactor overflow. Surprisingly, it has been found that this object can be achieved by means of a settling zone in the reactor. This settling zone is designed in such a way that it has a feed line for the mixture from below and is sufficiently large that the solid in the inflowing mixture can settle out.

Similar demands are made of the material of the downstream heatable tubular reactor as are made of the stirred reactor. Besides steel, nickel and nickel alloys, steel reactors coated with PTFE/PFA, enamel or ceramic materials are also suitable.

An essential advantage is the combination of stirred reactor with tubular reactor. A higher temperature can be set in the downstream tubular reactor. This results in undissolved alkali metal salt dissolving and reacting in the melt at the higher temperatures in a finite residence time.

The reaction in the stirred reactor and tubular reactor can be carried out in the presence of atmospheric oxygen or optionally under a protective-gas atmosphere (for example nitrogen, CO_2, noble gases) under reduced pressure, atmospheric pressure or even under superatmospheric pressure, at temperatures of from 50°C to 800°C (under atmospheric pressure). When working under superatmospheric pressure or reduced pressure, the melting points of the salts shift correspondingly.

The processing should be carried out at below the sublimation temperature of the starting materials. Preference is given to higher temperatures, since the solubility of the salts is significantly better under such conditions.

During the processing of the salts in the stirred reactor and in the tubular reactor, an optimum temperature programme can be set during the process by means of heater bands or twin-jacket heating.

The process can be carried out continuously or discontinuously, as required.

In order to carry out the process, the metal halide employed is aluminium, gallium, indium or thallium fluoride, chloride, bromide or iodide, or mixtures thereof. Suitable alkali metal salts are lithium, sodium, potassium, rubidium or caesium fluoride, chloride, bromide or iodide, or mixtures thereof.

A general example of the invention is explained in greater detail below and is shown in the drawing. Fig. 1 shows a stirred reactor 1 with solids metering devices 2 and 3, a tubular reactor 4, and downstream columns or towers 5 and 6.

In order to prepare salts conforming to the formula (I), and mixtures thereof, the starting materials can be fed to the reactor separately via solids metering devices (2) and (3). The starting materials can also be fed in, premixed, in the same ratio via a single metering device. The filling can also be carried out under an inert gas.

The heatable stirred reactor (1) with stirrer contains a liquid salt melt. The volume of this melt is determined depending on the required residence time and the desired throughput. The materials are reacted in the stirred reactor at temperatures above the melting point of the salt and below the sublimation temperature of the respective metal halide.

The fill level of the reactor is kept constant by means of an overflow pipe or an electrically controlled pump. In order that no solid starting material enters the overflow, the reactor contains a settling zone which has a feed line for the mixture from below and is sufficiently large that the solid can settle out in the outflowing mixture. This zone does not have a feed from the liquid surface and is sufficiently screened from the turbulence of the stirrer. Alternatively, use can be made of a rotating drum from whose centre the

product is discharged and whose centrifugal force separates off the solid. A hydrocyclone is also suitable for this task if an adequate flow rate is generated via a pump.

In order to increase the efficiency, a tubular reactor (4) is connected downstream of the stirred reactor.

The temperatures in the tubular reactor can be higher than in the stirred reactor. This enables undissolved alkali metal salt to be dissolved in the melt and the yield to be increased.

The tubular reactor is advantageously arranged vertically. This prevents deposition of the solid on the vessel wall. In addition, pumps are unnecessary since material transport takes place via gravity.

The melt may have been be contaminated by contact with water or atmospheric moisture. The hydrogen halide formed is passed through a column or tower (5) filled with the appropriate metal granules. The melt advantageously passes through the column or tower from bottom to top. The hydrogen halide reacts with the metal to form DX_3. For further processing, the metal halide is passed through a further column or tower (6). The flow through the column or tower charged with alkali metal salt MX is likewise from bottom to top. The metal halide DX_3 is reacted with the alkali metal salt MX here to form the desired salt MDX_4.

It is not vital that the flow through the columns or towers (5) and (6) is from bottom to top, but this does have the advantage that the metal particles D and metal halide particles DX_3, which are becoming smaller owing to the reaction, are not forced against the screen plate by the flow and block the latter. Nevertheless, homogeneous flow (plug flow) in the column is ensured

in this way. Homogeneous flow is an essential pre-requisite for complete reaction in the column.

The example given below is given in order to better illustrate the present invention, but is not suitable for restricting the invention to the features disclosed therein.

Examples

Example 1:

Preparation of $NaAlCl_4$

In order to prepare 1 kg/h of $NaAlCl_4$, 373.8 g/h of NaCl are fed to a heatable stirred reactor via a solids metering device and 626.2 g/h of $AlCl_3$ are fed to the reactor via a further solids metering device. The glass stirred reactor with propeller stirrer and oil-heated twin jacket contains a volume of liquid salt melt at a temperature which is below the sublimation temperature of the $AlCl_3$ (180°C), but above the melting point of the salt (156°C). Vigorous stirring causes intimate contact between the liquid melt and the heated reactor wall on one hand and the cold starting material on the other hand. This results in good heat transfer, and consequently the average residence time is about 5 minutes.

The fill level of the reactor is kept constant by means of an overflow pipe. Contamination of the products by unreacted starting materials is excluded here by the connection downstream of a tubular reactor (4). Due to the setting of elevated temperatures in the tubular reactor, here 200°C, undissolved NaCl can react in the melt. The vertically arranged tubular reactor prevents the deposition of the solid which remains. In addition, pumps are unnecessary since material transport takes place via gravity.

The melt possibly contaminated by water can form hydrogen halide. This can be removed by reaction in two downstream columns filled with aluminium granules (5) and with NaCl (6). In the columns, through which flow is from bottom to top, firstly the hydrogen halide can react with aluminium to give $AlCl_3$ and subsequently in

the second column with the salt NaCl to give the product $NaAlCl_4$.

PATENT CLAIMS

1. Apparatus for the preparation of salt melts, and mixtures thereof, of the general formula

$$MDX_4 \quad (I)$$

in which

M is Li, Na, K, Rb or Cs,

D is Al, Ga, In or Tl, and

X is F, Cl, Br or I,

essentially consisting of a heatable stirred reactor (1) and a downstream tubular reactor (4), where the stirred reactor (1) contains a zone which, owing to the tank geometry, cannot contain solids, and the tubular reactor (4) or its feed line extends into this solid-free zone.

2. Apparatus according to Claim 1, characterized in that the tubular reactor (4) is arranged vertically.

3. Apparatus according to Claim 1 or 2, characterized in that solids metering units (2, 3) for controlled addition or mixing of the starting materials are arranged upstream of the stirred reactor (1).

4. Apparatus according to one of Claims 1 to 4, characterized in that a purification unit (5, 6), consisting of a column or tower (5) filled with metal granules (D) and a column or tower filled with alkali metal salt (MX), is arranged downstream of the tubular reactor (4).

5. Process for the preparation of salt melts of the general formula

$$MDX_4 \quad (I)$$

in which

M is Li, Na, K, Rb or Cs,

D is Al, Ga, In or Tl, and

X is F, Cl, Br or I,

by reacting a metal halide of the formula DX_3 (II) with an alkali metal salt of the formula MX (III), characterized in that the reaction is carried out in an apparatus according to Claims 1 to 5, where the reaction is carried out firstly in a stirred reactor (1) and subsequently in a tubular reactor (4).

6. Process according to Claim 5, characterized in that the salts are reacted at different temperatures in the stirred reactor (1) and the tubular reactor (4).

7. Process according to Claim 5 or 6, characterized in that the salts are reacted at temperatures between 50 and 800°C.

8. Process according to one of Claims 5 to 7, characterized in that the reaction is carried out continuously.

9. Use of salts of the formula (1), defined in Claim 1 and prepared by a process according to Claims 5 to 8, as melt electrolyte in electrochemical cells, batteries, sodium batteries and primary batteries.

10. Use of salts of the formula (1), defined in Claim 1 and prepared by a process according to Claims 5 to 8, as storage medium in heat stores and as heat-transfer medium.

11. Use of salts of the formula (1), defined in Claim 1 and prepared by a process according to Claims 5 to 8, for blanketing and purifying molten metals and for electrocoating materials.

Docket No.
Merck

Declaration and Power of Attorney For Patent Application
English Language Declaration

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name,

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

Apparatus and process for the preparation of salt melts, and the use thereof

the specification of which

(check one)

☐ is attached hereto.
☒ was filed on 08.03.2000 as United States Application No. or PCT International Application Number PCT/EP00/02009
and was amended on ______
(if applicable)

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose to the United States Patent and Trademark Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, Section 1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, Section 119(a)-(d) or Section 365(b) of any foreign application(s) for patent or inventor's certificate, or Section 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate or PCT International application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application(s)			Priority Not Claimed
199 13 190.2 (Number)	Germany (Country)	24.03.1999 (Day/Month/Year Filed)	☐
(Number)	(Country)	(Day/Month/Year Filed)	☐
(Number)	(Country)	(Day/Month/Year Filed)	☐

I hereby claim the benefit under 35 U.S.C. Section 119(e) of any United States provisional application(s) listed below:

(Application Serial No.)	(Filing Date)
(Application Serial No.)	(Filing Date)
(Application Serial No.)	(Filing Date)

I hereby claim the benefit under 35 U.S.C. Section 120 of any United States application(s), or Section 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States of PCT International application in the manner provided by the first paragraph of 35 U.S.C. Section 112. I acknowledge the duty to disclose to the United States Patent and Trademark Office all information known to me to be material to patentability as defined in Title 37, C.F.R., Section 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application:

(Application Serial No.)	(Filing Date)	(Status) (patented, pending, abandoned)
(Application Serial No.)	(Filing Date)	(Status) (patented, pending, abandoned)
(Application Serial No.)	(Filing Date)	(Status) (patented, pending, abandoned)

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

POWER OF ATTORNEY: As a named inventor, I hereby appoint

I. William Millen (Reg. No. 19,544)
John L. White (Reg. No. 17,746)
Anthony J. Zelano (Reg. No. 27,969)
Alan E.J. Branigan (Reg. No. 20,565)
John R. Moses (Reg. No. 24,983)
Harry B. Shubin (Reg. No. 32,004)
Brion P. Heaney (Reg. No. 32,542)
Richard J. Traverso (Reg. No. 30,595)
John A. Sopp (Reg. No. 33,103)
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